PB82-118159

Mobile Source Emissions of Formaldehyde and Other Aldehydes

(U.S.) Environmental Protection Agency
Ann Arbor, MI

May 81

U.S. Department of Commerce
National Technical Information Service
NTIS.

PB82-118159

EPA/AA/CTAB/PA/81-11

MOBILE SOURCE EMISSIONS OF FORMALDEHYDE AND OTHER ALDEHYDES

by

Penny M. Carey

May 1981

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Ann Arbor, Michigan 48105

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SPRINGFIELD, VA 22161

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
EPA-AA-CTAB-PA-81-11		PB82 118159

4. TITLE AND SUBTITLE	5. REPORT DATE
Mobile Source Emissions of Formaldehyde and other Aldehydes.	May 1981
	6. PERFORMING ORGANIZATION CODE 306002
7. AUTHOR(S) Penny M. Carey	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS U.S. Environmental Protection Agency Office of Mobile Source Air Pollution Control Control Technology Assessment & Characterization Branch Ann Arbor, Michigan 48105	10. PROGRAM ELEMENT NO.
	11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS SAME AS BOX 9	13. TYPE OF REPORT AND PERIOD COVERED Technical
	14. SPONSORING AGENCY CODE

15. SUPPLEMENTARY NOTES

16. ABST

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17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Formaldehyde Aldehydes Automobiles		

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)*	21. NO. OF PAGES
release unlimited	unclassified	35
	20. SECURITY CLASS *(This page)* unclassified	22. PRICE

EPA Form 2220-1 (9-73)

Mobile Source Emissions of Formaldehyde and Other Aldehydes

I. Introduction

Aldehydes are a class of partially oxidized hydrocarbons emitted from many sources, including mobile sources. Vehicular aldehyde emissions are currently unregulated; however, preliminary results of a 24-month inhalation study indicate that formaldehyde, the most prevalent aldehyde in vehicular emissions, is a carcinogen in rats. The results of this study may serve as an impetus for future regulatory action and possible classification of formaldehyde as a human carcinogen. Consequently, there has been an increasing number of studies examining the potential health risks and relative contribution of the aldehydes as exhaust pollutants. Topics covered in these studies include aldehyde emission factors for unmodified and malfunction vehicle engine configurations, effects of mileage accumulation, fuel and temperature variations and aldehyde emissions from Diesel-equipped vehicles equipped with prototype light-duty Diesel oxidation catalysts. The purpose of this document is to summarize these studies and formulate formaldehyde and total aldehyde emission factors for each vehicle class.

II. Summary

The available vehicular aldehyde studies were summarized in an attempt to characterize aldehyde emissions from motor vehicles. Topics covered in these studies include aldehyde emission factors for unmodified and malfunction vehicle engine configurations, effects of fuel, mileage accumulation and temperature variations and aldehyde emissions from Diesel-equipped vehicles equipped with prototype light-duty Diesel oxidation catalysts. Thus, it was possible to obtain aldehyde data for standard conditions and for a variety of operating conditions. The Federal test procedure (FTP) was used for the light-duty vehicles and the 13-mode test procedure for the heavy-duty engines. The 2, 4 dinitrophenylhydrazine (DNPH) procedure was used for the sampling and analysis of the aldehydes. This procedure is discussed in the Appendix. In addition to aldehydes, the DNPH procedure detects two ketones, methylethylketone and acetone. Methylethylketone measurements are not included in this report. However, acetone and two aldehydes, acrolein and propionaldehyde are reported together as acetone since they are not resolved from each other under normal gas chromatographic operating conditions. The term "total aldehydes", as used in this report, includes the acetone measurements.

Based on the studies summarized, the following generalizations can be made:

1. For the light-duty vehicles, highest average total aldehyde emissions occurred with the gasoline fueled non-catalyst-equipped vehicles (88 mg/km) followed by the Diesel engine-equipped vehicles (37 mg/km) and the gasoline fueled catalyst-equipped vehicles (2 mg/km). Formaldehyde emissions showed a similar trend. On the same basis (mg/km), the heavy-duty vehicles, in turn, emitted more formaldehyde and total aldehydes than the light-duty vehicles.

2. Results for the malfunction configuration studies involving light-duty gasoline fueled vehicles indicate that, for the non-catalyst-equipped vehicles and vehicles equipped with oxidation catalysts, aldehyde emissions tend to increase most during 12 percent misfire (by factors of 9 and 13, respectively compared to the unmodified configuration). Aldehyde emissions for these vehicles also increase slightly during rich best idle and disabled EGR malfunction configurations. The results for the three-way catalyst-equipped vehicles were not as conclusive. The greatest number of three-way catalyst-equipped vehicles for which the emissions increased occurred during a disabled O_2 sensor malfunction. The catalyst-equipped vehicles appear to be more effective in controlling THC, formaldehyde and total aldehyde emissions than non-catalyst-equipped vehicles, even under malfunction conditions.

3. A Diesel fuel study was conducted with light-duty vehicles and heavy-duty engines. For the light-duty vehicles, total aldehyde and formaldehyde emissions were generally the lowest when "Jet A" No. 1 fuel was used and highest when the Minimum Quality No. 2 fuel was used. Minimum and maximum total aldehyde emissions differed by as much as a factor of 4. THC emissions behaved similarly. There was more variability for the heavy-duty engines, compared to the light-duty vehicles, and few trends were apparent.

4. The results of a temperature study were variable, but in general aldehyde emissions tend to increase as the temperature decreases. The total range for the aldehyde emissions varied from 19 mg/km for a 1980 Buick prototype (soaked and tested at 80°F with the air conditioner operating) to 285 mg/km for a 1977 Honda Civic without a catalyst (soaked and tested at 20°F).

5. A 1979 Oldsmobile Diesel Delta 88 was used to evaluate emissions from prototype catalytic converters for Diesel exhaust supplied by two manufacturers, Englehard and UOP, Inc. A total of three catalysts were evaluated; one fresh Engelhard catalyst, one fresh UOP catalyst and an aged UOP counterpart (subjected by UOP to 81,000 km of road aging on a similar vehicle before delivery to the test site). The vehicle was also tested without a catalyst. The fresh catalysts were found to be effective in decreasing the amount of aldehydes and THC in the exhaust; however, the fresh UOP catalyst exhibited substantially greater activity for reducing THC and aldehydes than the aged UOP counterpart. Average total aldehyde emissions for the fresh Engelhard catalyst were 2.5 mg/km compared to 6.2 mg/km for the non-catalyst baseline. Average total aldehyde emissions for the fresh UOP catalyst were 4.3 mg/km compared to 5.7 mg/km for the aged counterpart. Average THC emissions for the fresh Engelhard catalyst, fresh UOP catalyst, aged UOP catalyst and non-catalyst baseline were 90, 85, 195 and 245 mg/km respectively.

6. The effect of mileage accumulation on aldehyde emissions from gasoline fueled catalyst-equipped vehicles has not been extensively studied. General Motors has conducted tests on four high mileage vehicles equipped with oxidation catalysts. Odometer readings ranged from 69,417 to 76,099 miles. Aldehyde emissions from these high mileage vehicles

averaged 18.05 mg/km in comparison to 12.4 mg/km for low mileage catalyst-equipped vehicles and 87 mg/km for non-catalyst-equipped vehicles (all tested by GM).

In an EPA sponsored study, aldehyde emissions from four new three-way catalyst-equipped vehicles were evaluated initially and at three mileage accumulation points (5000 mile increments). While some of the vehicles did not show a continuous increase in aldehyde emissions with mileage, they all showed an increase in individual and total aldehydes from the initial to the final mileage test point. In an attempt to determine if this trend continues as mileage increases, the data from this study were extrapolated to higher mileages (50,000 + miles) using linear regression. Deterioration factors were then calculated at selected high mileage points and used with low mileage aldehyde data from oxidation catalyst-equipped vehicles to project aldehyde emissions from high mileage oxidation catalyst-equipped vehicles. These projected emissions were then compared to the actual high mileage aldehyde data generated by GM. The projected values for aldehydes and THC exceeded the values obtained by GM. It appears that aldehyde and THC emissions do not increase at a constant rate with increasing mileage; however, it is not certain whether, 1) these emissions level off at some point, or 2) continue to increase with mileage but at a slower rate than indicated by the study. In either case, the projected high mileage aldehyde emissions from catalyst-equipped vehicles were less than aldehyde emissions found from non-catalyst-equipped vehicles. An EPA sponsored study is presently underway to investigate aldehyde emissions from high mileage catalyst-equipped vehicles.

III. Definition/Sources

Aldehydes are a chemical class of partially oxidized hydrocarbons. A partially oxidized hydrocarbon is a hydrocarbon containing one or more oxygen molecules in addition to hydrogen and carbon. The partially oxidized hydrocarbon with the chemical formula HCHO is known as formaldehyde and its structure is shown below.

formaldehyde

In general, aldehydes are a chemical class of partially oxidized hydrocarbons that contain the group -CHO. Table 1, at the end of the text, contains a list of a few of the aldehydes, including those measured in motor vehicle exhaust, along with their corresponding chemical structures.

Formaldehyde is one of the primary aldehydes of importance in industrial use. Several billion pounds of formaldehyde are produced each year in the United States. Partially because of formaldehyde's antiseptic properties, it is used in the medical, brewing, and agricultural industries. It is also widely used in the manufacture of phenolic, urea, and melamine resins.

Under certain conditions, formaldehyde can be released into the environment over a prolonged period from resinous products. These products include ureaformaldehyde foam insulation, particle board, and even some plywoods. Additional sources of formaldehyde include automotive exhaust, smog formed in the ambient air, cigarette smoke and incinerators. Aldehydes and formaldehyde are often formed by the incomplete combustion of hydrocarbons. Aldehydes in mobile source exhaust are formed by the incomplete combustion (partial oxidation) of the fuel.

IV. Health Effects

This section summarizes some of the available health information on aldehydes. The aldehydes have a characteristically pungent odor, are flammable, are photochemically reactive, can cause respiratory problems and are eye irritants. The two major aldehydes of interest from a human health perspective appear to be formaldehyde and acrolein. Both are severe eye irritants. In addition, exposure to formaldehyde causes upper respiratory tract irritation, headaches, gastrointestinal problems and skin irritation.

Formaldehyde is known to be a component of photochemical smog formation. Photochemical smog is a form of air pollution which arises from the reactions of oxides of nitrogen and hydrocarbon compounds in the presence of sunlight. Formaldehyde can be photooxidized with a nitrogen oxide mixture in air to yield ozone, which is also toxic. Smog often results in eye and throat irritation, odor, plant damage and decreased visibility. Formaldehyde may account for a large fraction of the eye irritation associated with photochemical air pollution.

There are no published data to indicate that formaldehyde is a confirmed carcinogen in humans. However, preliminary results of a 24-month inhalation study sponsored by the Chemical Industry Institute of Toxicology (CIIT) in Research Triangle Park, North Carolina indicate that formaldehyde is a carcinogen in rats. After 16 months, three male rats exposed to a formaldehyde level of 15 ppm for six hours per day, five days per week were found to have squamous cell carcinoma of the nasal passages. The frequency of nasal tumors through the 18 month sacrifice was reported by Swenberg et al. (1)*. These results provided evidence that formaldehyde might represent a carcinogenic risk to humans.

In April, 1980, a panel of scientists from within the Federal Government was formed under the guidance of the National Toxicology Program and coordinated by the Consumer Product Safety Commission. The panel members reviewed and evaluated the available published and unpublished information on the adverse health effects from repeated exposure to formaldehyde. Acute toxic effects and hypersensitivity were not considered by the panel since they had recently been assessed by the Committee on Toxicology of the National Academy of Sciences (2). The Panel released their report in November, 1980 (3). The Panel concluded that definitive experiments exist which demonstrate the mutagenicity and carcinogenicity of formaldehyde under laboratory conditions. The Panel also concluded that formaldehyde should be presumed to pose a carcinogenic risk to humans.

V. Methods of Measurement

Two major methods have been used for the sampling and analysis of aldehydes in vehicle exhaust, the 3-methyl-2-benzothiazolone hydrazone (MBTH) method (4) and the 2, 4 dinitrophenylhydrazine (DNPH) procedure (5). The MBTH method detects total aliphatic aldehydes and is not capable of separating individual components. The MBTH method has been found to give generally low results due to a positive interference of oxides of sulfur on the absorption readings. Previous data from an EPA-DOE program at Bartlesville (6) indicate that the DNPH method gave, on the average, 44 percent higher response than the MBTH method. Unlike the MBTH method, the DNPH method detects individual aldehydes and ketones. The individual aldehydes and ketones that are included in this analysis are: formaldehyde, acetaldehyde, acetone, acrolein, propionaldehyde, isobutyraldehyde, methylethylketone, crotonaldehyde, hexanaldehyde and benzaldehyde. A summary of the DNPH procedure can be found in the Appendix. Phthalates and di-2-ethylhexyladipate were found by mass spectroscopy to be interferences in the procedure (5). Many phthalate esters are found in lubricants and plastics, and di-2-ethylhexyladipate is used as a fuel stabilizer. The crotonaldehyde and benzaldehyde values can be affected by these interferences. The DNPH procedure is also labor intensive but is capable of at least a 98 percent collection efficiency.

The emission factor data reported here were found using the DNPH method (with gas chromatograph/flame ionization detector analysis) unless otherwise stated. Mention should be made of the two ketones detected with this procedure, methylethylketone and acetone. Methylethylketone measurements are not included in this report. However, acetone and two aldehydes, acrolein and propionaldehyde are reported together as acetone since they are not resolved from each other under normal gas chromatographic operating conditions. The term "total aldehydes", as used in this report, includes the acetone measurements. It should be noted that work is currently being performed to improve both the MBTH (7) and DNPH (8,9) procedures.

VI. Summaries of Aldehyde Studies and Emission Factor Data

A. Manufacturers' Status Reports

At the request of the EPA, the manufacturers submit status reports summarizing their emission control development and testing programs. Many of the manufacturers have measured unregulated emissions as part of their testing program. The following is a summary of some of the manufacturers' research with regard to the collection and measurement of aldehydes in vehicle exhaust (10).

GM has presented data on total aldehyde emissions using the MBTH method and 1975 FTP for two Diesel-equipped vehicles, a 1976 Mercedes 300D and a 1976 Peugeot 504D. The results were 13.7 mg/km and 37.3 mg/km respectively.

*Numbers in parentheses designate references at end of paper.

The MBTH method and 1975 FTP were used to measure total aldehyde emissions for two cars as part of an evaluation of alcohol-based motor fuels. The first car was a Brazilian Chevrolet Opala with no emission controls operated on gasoline/ethanol blends. The HC, CO and NOx emissions were 4.8, 74.5 and 0.8 g/km respectively. The aldehyde emissions were 33.6 mg/km on gasoline, 42.3 mg/km on 5% ethanol, 59 mg/km on 10% ethanol and 75.8 mg/km on 20% ethanol. The second car was a 1974 Chevelle operated on pure methanol. The aldehyde emissions were 181.5 mg/km at one spark carburetor setting and 121.8 mg/km at another. The car was not tested on gasoline.

Other data on GM aldehyde work was found in a Feb. 1, 1978 letter from T. M. Fisher (GM) to Dr. James N. Pitts (University of California). These data, according to GM, show that aldehydes are reduced in automotive exhaust by catalysts even under adverse oxidation conditions and after the normal decrease in hydrocarbon oxidation efficiency has occurred with mileage. The results of GM tests conducted on four high mileage vehicles equipped with oxidation catalysts are shown in Table 2. Also shown as part of Table 2 is a comparison of the range of the aldehyde emissions for non-catalyst-equipped vehicles, low and high mileage catalyst-equipped vehicles and Diesel-equipped vehicles. Aldehyde emissions from non-catalyst-equipped vehicles averaged 87 mg/km in comparison to 12.4 mg/km for low mileage catalyst-equipped vehicles, 18.05 mg/km for high mileage catalyst-equipped vehicles and 24.85 mg/km for Diesel-equipped vehicles (all tested by GM). Table 2 also includes exhaust aldehyde emissions of special cars (11). GM concludes that the catalyst is generally more effective in controlling aldehyde emissions than it is for total hydrocarbons.

Ford focused its attention on evaluation of aldehyde measurement procedures. A revised MBTH method was evaluated for the determination of low concentrations of total aliphatic aldehydes in exhaust. The revised procedure is not subject to interferences encountered in earlier MBTH methods.

Ford has experimented with the DNPH method for determination of total aldehydes and developed an oxime technique for determination of airborne aldehydes. In addition, a gas analysis system composed of a Fourier Transform Infra-Red (FTIR) spectrometer, a quadrupole mass spectrometer and a total hydrocarbon analyzer has been developed. Both regulated and unregulated emissions, including individual aldehydes, can be measured with this gas analysis system.

Ford has measured ethanol and total aliphatic aldehyde emissions from a 100% ethanol fueled Corcel II vehicle. Samples were taken from undiluted raw exhaust and exhaust diluted in the constant volume sampling (CVS) test. Runs were conducted with and without a copper alumina oxidation catalyst. Over the duration of the CVS test, aldehyde emissions were about 25% higher with the catalyst than without it. Similar results were found for the undiluted raw exhaust; the average aldehyde emission increase was less than 8% with the catalyst. It is possible that the unburned ethanol in the exhaust was only partially oxidized by the catalyst, resulting in increased aldehyde emissions.

Leyland used the MBTH method to report total aldehyde emission rates for three Triumphs. They were 6.8 mg/km (over hot LA-4) for a 1977 Triumph TA7-2V (Federal emission standards), 4.8 mg/km (over cold LA-4) for a 1977 Triumph TA7-2V (California emission standard) and 4.0 mg/km (over hot LA-4) for a 1978 Triumph Spitfire 1500 (Federal emission standards).

Honda reported total aldehyde emissions for a CVCC engine (19.6 mg/km) and a conventional engine (30.2 mg/km) at 25 MPH steady state using the DNPH method. Honda is also conducting studies using a high performance liquid chromatograph for measurement of individual aldehydes.

Toyo Kogyo (Mazda) measured aldehyde emissions from a 1978 Mazda RX-3 equipped with a thermal reactor and a 1978 GLC equipped with an oxidation catalyst. The tests used were FTP, 0-2 min (choke was pulled out fully when starting, pushed in to a half setting in 5 seconds and pushed in fully in another 2 minutes), 2-10 min (after cold starting) and idle (after sufficient idling). The test results indicated lower emission rates for the 1978 GLC with an oxidation catalyst regardless of the test used.

In a later study, Toyo Kogyo measured aldehyde emissions from four gasoline vehicles equipped with catalysts. The maximum emission rates found for five individual aldehydes were compared to an exposure level and an evaluation standard determined for each individual aldehyde. The exposure levels for each aldehyde were the 24-hour exposure levels on a highway assuming that all vehicles emit the maximum emission rates of each aldehyde. The evaluation-standard exposure levels at roadside locations of a city were set at one-hundredth of the ACGIH-TLV* for each aldehyde, in consideration of infirm persons, children, etc. and also because people are exposed to the pollutants for a long time. The exposure levels of the aldehydes were greatly lower than the evaluation-standard levels in spite of the severe conditions assumed.

VW presented total aldehyde data for eight gasoline VW vehicles, five equipped with catalysts. The MBTH method was used. These data, shown in Table 3, generally indicate a substantial decrease in aldehyde emissions of catalyst-equipped vehicles over the non-catalyst-equipped vehicles. VW also measured total aldehyde emissions from a naturally aspirated 50 hp, a turbocharged 70 hp and a naturally aspirated 66 hp Diesel. The aldehyde emission levels for the Diesel and gasoline vehicles were comparable.

Nissan is presently using the MBTH method but plans to use the DNPH method in the future. Nissan has measured aldehyde emissions from both gasoline and Diesel-equipped vehicles. A synopsis of this research follows.

Diesel testing was conducted to determine the effect the presence or absence of EGR had on unregulated emissions. When NOx levels are lowered using EGR, the emission levels of aldehydes rise markedly, according to Nissan. When

*American Conference of Governmental Industrial Hygienists-Threshold Limit Value

emissions from gasoline and Diesel-equipped vehicles were compared, Nissan found that aldehyde emissions from Diesel-equipped vehicles are the same as aldehyde emissions from gasoline vehicles without catalytic converters but are a factor of 10 greater than gasoline vehicles equipped with catalytic converters.

Nissan has examined the effect of mileage accumulation on unregulated emissions using vehicles equipped with catalysts which had been put through an AMA 50,000 mile durability run. Components such as aldehydes and hydrocarbons that are originally present in the exhaust and are reduced by the catalyst show a tendency to increase after the mileage accumulation.

Methanol (15 vol. %) was blended into the gasoline to investigate the effect of an alternate fuel on aldehyde emissions. With the methanol blend, the aldehyde emissions increase from two to five times in comparison with gasoline. In an AMA 30,000 mile durability test using this blend, there was no particular change in the aldehyde emissions.

Nissan has also investigated how various malfunctions (air-fuel ratio, secondary air induction system and EGR) would affect the unregulated emissions from a vehicle equipped with an oxidation catalyst. When the air-fuel ratio is on the rich side, aldehydes decrease since they are the product of an oxidizing atmosphere. When the air-fuel ratio deviates on the lean side, the aldehydes do not show any significant change. A secondary air induction system (EAI) malfunction results in a reducing atmosphere; consequently, aldehyde emissions decrease. The aldehydes did not appear to be affected by an EGR system malfunction.

In general, emissions of aldehydes have been shown to decrease when a catalyst is used for emission control. Control of HC and CO emissions brings about a corresponding reduction in aldehyde emissions for most emission control systems.

B. Light-Duty Vehicle Aldehyde Emissions

1. Light-Duty Gasoline

The aldehyde emission factors for a few light-duty catalyst-equipped and non-catalyst-equipped gasoline vehicles are shown in Tables 4 and 5, respectively. Data for the catalyst-equipped vehicles come from work performed by Southwest Research Institute under four contracts to the Environmental Protection Agency (12, 13, 14, 15). Data for the non-catalyst-equipped vehicles were taken from two EPA sponsored contracts (6, 16).

The aldehyde emissions for all light-duty vehicles were measured by the DNPH method. Values for crotonaldehyde and benzaldehyde are not included in the tables due to the aforementioned interferences with the DNPH procedure. The light-duty Federal Test Procedure (FTP) was used.

For the catalyst-equipped vehicles, total aldehyde values ranged from 0.375 mg/km (1978 Saab 99) to 6.80 mg/km (1977 Olds Cutlass) with an average emission factor of 2.36 mg/km. The average formaldehyde emission factor is 1.26 mg/km. On a mass basis, total aldehyde emissions are roughly 1.3

percent of the total hydrocarbon (THC) emissions for the catalyst-equipped vehicles. These averages have been computed using vehicles equipped with oxidation catalysts, three-way catalysts and three-way plus oxidation catalysts. The three-way catalysts appear to be the most effective for controlling formaldehyde and total aldehyde emissions. The addition of an oxidation catalyst to a three-way catalyst system, however, results in increased formaldehyde and total aldehyde emissions, approaching levels characteristic of operation with an oxidation catalyst alone. Average emission factors are presented in Table 4 computed with and without the inclusion of three-way and three-way plus oxidation catalyst-equipped vehicles. The average emission factors for the aldehydes and THC decrease when the three-way and three-way plus oxidation catalyst-equipped vehicles are included. It should be noted when reviewing these data that the catalyst-equipped vehicles used for this analysis were low mileage vehicles tuned to manufacturers' specifications.

Table 5 presents data for the non-catalyst-equipped vehicles. As seen in Table 5, individual aldehyde data are not available for 10 of the vehicles. These vehicles were part of a program designed to characterize aldehyde and reactive organic emissions from 1970 through 1973 model vehicles. Odometer readings varied froom 2288 to 68,779 miles. Unfortunately, since the aldehyde distribution in the exhaust of these vehicles is unknown, it is not possible to determine if some of the unusually high values for total aldehydes (e.g. 205 mg/km for the 1970 Pontiac) are due to interferences with the sampling procedure. Individual aldehyde data are available for four 1970 vehicles. These vehicles are part of a recent effort to determine unregulated exhaust emissions from non-catalyst baseline vehicles. The vehicles were 10 years old upon receipt and were tuned to manufacturers' specifications prior to testing.

The total aldehyde average emission factor for the non-catalyst-equipped vehicles was 88 mg/km, much higher than the catalyst-equipped vehicles but not unexpected. The formaldehyde average emission factor was 32.31 mg/km, over 20 times that of the catalyst-equipped vehicles. (This figure is based on four vehicles.) On a mass basis, total aldehyde emissions are roughly 4.2 percent of the THC emissions for the non-catalyst-equipped vehicles.

2. Light-Duty Diesel

The aldehyde emission factors for some light-duty Diesel-equipped vehicles can be found in Table 6. The data in Table 6 were generated in two EPA sponsored studies (12, 17). Total aldehyde emission values ranged from 8.75 mg/km (1975 Mercedes 240D) to 76.50 mg/km (1976 Cutlass Diesel) with an average of 36.70 mg/km. The average formaldehyde emission rate for the light-duty Diesels is 13.07 mg/km, falling between the values given for the catalyst-equipped vehicles (1.26 mg/km) and the non-catalyst-equipped vehicles (32.31 mg/km). Total aldehyde emissions are roughly 11.5 percent of the THC emissions.

3. Summary - Light-Duty Vehicles

The light-duty gasoline catalyst-equipped vehicles had the lowest average formaldehyde and total aldehyde emission rates. These values may be

especially low due to the inclusion of three-way catalyst-equipped vehicles. Cars equipped with dual or three-way catalyst systems have lower aldehyde emissions (see Tables 2 and 4). The non-catalyst-equipped gasoline vehicles produced the highest average formaldehyde, and total aldehyde emission rates; however, the light-duty Diesel-equipped vehicles produced the largest percentage of aldehydes in the total hydrocarbon exhaust. It is evident that aldehyde emissions could constitute an appreciable fraction of the photochemical oxidant potential of Diesel exhaust.

C. Heavy-Duty Engine Aldehyde Emissions

Aldehyde emission factors for three heavy-duty engines are listed in Table 7. The data reported here come from work performed by Southwest Research Institute under contract to EPA (12). The DNPH method was used to measure the heavy-duty engine aldehyde emissions. The 13-mode FTP (steady state) test procedure was selected. Two Diesel heavy-duty engines were tested, a Mack ETAY(B)673A and a Caterpillar 3208/EGR. The Chevrolet 366 engine was chosen to represent the gasoline heavy-duty engine. Both the Mack and the Caterpillar used No. 2 Diesel fuel, representative of "national average" properties. The Chevrolet engine used leaded gasoline. The aldehyde emissions are listed in units of g/kw-hr, g/kg fuel and g/km. The work specific rates (g/kw-hr) and fuel specific emission rates (g/kg fuel) were taken from the EPA contract project (12). The emission factors (g/km) were derived using the fuel specific emission rates and information taken from the article "Heavy-Duty Diesel Particulate Emission Factors" (18). The emission factors derived are crude approximations and are included as a means of comparison with other vehicle types. The conditions used in formulating these emission factors included half load and combined city-highway driving. The emission factors used were the average of the New York City and Los Angeles derived emission factors. These conditions represent an estimate of the general population's exposure to aldehyde emissions from heavy-duty vehicles.

On a brake specific basis, the Chevrolet 366 gasoline engine emitted more aldehydes overall. Formaldehyde emissions were substantially higher. Crotonaldehyde and benzaldehyde emissions given in the original report were extremely high and are probably the result of artifacts in the sampling procedure; therefore,they were not included in Table 7. Total aldehyde emissions are roughly 5.9 percent of the THC emissions for the gasoline engine versus roughly 7.8 percent for the Diesel-equipped heavy-duty engines.

An overall summary of the aldehyde and hydrocarbon emissions for the light-duty vehicles and heavy-duty engines can be found in Table 8. It should be noted that the vehicles/engines used for this analysis were all tuned to manufacturer specifications.

D. Malfunction Configuration Studies - LD Gasoline Vehicles

Three malfunction configuration studies will be summarized here (13, 14, 15). A total of nine gasoline vehicles were tested in the unmodified configuration and in four engine and/or emission control system malfunction configurations; the results for six of the vehicles will be reported here.

The DNPH method and four cycle FTP testing procedures were used in each study. The specific malfunction configurations varied somewhat from vehicle to vehicle. One vehicle tested, a 1977 AMC Pacer, had no catalyst. Of the remaining five catalyst vehicles, one had an oxidation catalyst, two had three-way catalysts and two had three-way plus oxidation catalysts. The results are listed in Tables 9 and 10. The non-catalyst AMC Pacer produced the highest levels of formaldehyde and aldehydes in both the unmodified and malfunction configurations. The highest levels occurred at the 12 percent misfire condition (12 percent misfire was estimated to be the limit at which driveability remained reasonably acceptable to some people). Similar results have been obtained in a study using four 1970 non-catalyst-equipped vehicles (16). The Chevrolet Malibu behaved similarly with the highest levels of formaldehyde and total aldehydes occurring during the 12 percent misfire condition. The high oil consumption condition increased formaldehyde and total aldehyde levels for the non-catalyst-equipped vehicles but exhibited an inverse effect for the catalyst-equipped vehicles. The disabled EGR condition increased aldehyde emissions for both the catalyst-equipped and non-catalyst-equipped vehicles. The rich best idle increased the aldehyde emissions slightly for both vehicles tested.

The 1978 Saab 99 had the lowest unmodified aldehyde emissions of all the vehicles tested. Highest values for the Saab 99 occurred during the disabled O_2 sensor and rich best idle configurations. In general, the catalyst-equipped vehicles appear to be more effective in controlling THC, formaldehyde and total aldehyde emissions than non-catalyst-equipped vehicles, even under malfunction conditions.

The results listed in Tables 9 and 10 are average values. The unmodified values were obtained by averaging four runs while the malfunction configuration values were obtained by averaging two runs. The emission rates varied quite substantially in some cases from run to run. By averaging the runs the variability was reduced but the maximum values were lost.

E. Effects of Fuel Composition on Aldehydes

EPA has run several programs with various types of Diesel fuels. These data give an indication of how Diesel fuel composition affects aldehyde emissions.

The aldehyde emission factors for two light-duty and two heavy-duty Diesel equipped engines operated on five different fuels can be found in Table 11. The test fuel descriptions are as follows:

EM-238-F:	2D Emissions - "wide boiling range" fuel
EM-239-F:	National Average No. 2
EM-240-F:	"Jet A" No. 1
EM-241-F:	Minimum Quality No. 2 - a low-cetane (e.g. 42) high-aromatic Diesel fuel
EM-242-F:	Premium No. 2 - a high-cetane (e.g. 52) high paraffin Diesel fuel

The two light-duty Diesel-equipped vehicles tested were a Mercedes 240D and a VW Rabbit Diesel (19). The two heavy-duty Diesel-equipped engines tested were a Detroit Diesel Allison 6V-71 and a Caterpillar 3208 (20).

The VW Rabbit Diesel produced somewhat higher total aldehydes than the Mercedes 240D. Few trends seem to be evident regarding fuel effects. In both cases, however, the highest formaldehyde and total aldehyde values occurred when the Minimum Quality No. 2 fuel was used. Maximum and minimum total aldehyde emissions for the VW Rabbit differed by a factor of 2; for the Mercedes 240D by a factor of 4. THC emissions behaved similarly for both vehicles.

More variability is apparent in the heavy-duty 6V-71 data than in the 3208 data. The variability, according to the study, may be due to lubricating oil in the exhaust that created an interference with the analysis.

F. Temperature Study

It is known that emissions vary with ambient temperature. EPA has conducted some emission studies at various ambient temperatures. The composite aldehyde results of an ambient temperature study are shown in Table 12 (21). Fourteen vehicles were tested; three non-catalyst, three 49 state approved, five California approved and three prototype vehicles. Odometer readings ranged from 2774 miles (1978 Buick Regal) to 65,212 miles (1972 Chevrolet Impala). A modification of the DNPH method with colorimetric detection was used for determination of total aldehydes.

The 1974 Chevrolet Impala and 1977 Honda Civic, non-catalyst-equipped cars, showed the greatest aldehyde emissions. Air conditioning generally showed greater aldehyde emissions at 80 and 110°F (27 and 43°C) but less at 90°F (32°C). The total range for the values varied from 19 mg/km in the 1980 Buick prototype (soaked and tested at 80°F with air conditioning) to 285 mg/km in the Honda (soaked and tested at 20°F).

G. Prototype LD Diesel Catalyst Study

A preliminary investigation of prototype light-duty Diesel oxidation catalysts was performed (22). A 1979 Oldsmobile Diesel Delta 88 was used to evaluate emissions from prototype Diesel catalytic converters supplied by two manufacturers, Engelhard and UOP, Inc. The vehicle's exhaust system was modified twice in order to accomodate each of the manufacturer's systems. The vehicle was tested with and without a converter. A total of three catalytic converters were evaluated, two fresh and one aged. The Engelhard converter was a fresh monolithic oxidation catalyst and identified as PTX-516. There were two UOP monolithic oxidation catalysts identified as UOP-99 and UOP-103. Both were identical with the exception that UOP-99 was subjected to 81,000 km (50,342 miles) of road aging on a similar vehicle before delivery to the test site. The aldehydes were measured using the DNPH method. The Federal Test Procedure (FTP) results are shown in Table 13.

The reduction of the total amount of aldehydes for the Englehard catalyst was approximately 55 percent. Formaldehyde reduction was no greater than

the other aldehydes. Total hydrocarbons were reduced by approximately 60 percent.

The aldehyde and formaldehyde emissions for both UOP fresh and aged catalysts did not change appreciably from the non-catalyst baseline. However, the fresh catalyst exhibited substantially greater activity for reducing total hydrocarbons than its aged counterpart. The fresh UOP catalyst (UOP-103) reduced total hydrocarbons by approximately 50 percent. The aged UOP catalyst (UOP-99) reduced hydrocarbons approximately 30 percent.

H. Mileage Accumulation Study

A study was undertaken to characterize regulated and unregulated exhaust emissions at various mileages from three-way catalyst-equipped gasoline automobiles (23). In this study, four new three-way, catalyst-equipped automobiles were tuned to the manufacturer's specifications, and the emissions were evaluated over four test cycles with three unleaded gasolines initially and at three mileage accumulation points (5000 mile increments). The three fuels were: fuel 1, a low sulfur content fuel (less than 0.003 weight percent sulfur); fuel 2, fuel 1 blended to 0.03 weight percent sulfur, and fuel 3, a commercial grade fuel blended to 0.03 weight percent sulfur. A total of eight aldehydes and ketones were measured; however, the emission rate of two of the aldehydes, crotonaldehyde and benzaldehyde, could not be measured accurately due to interferences in the analysis procedure. Two other aldehydes and ketones, acetone and methylethylketone were found in only a small fraction of the samples analyzed. The FTP average emission rates for the remaining four aldehydes, formaldehyde, acetaldehyde, isobutyraldehyde and hexanaldehyde at each 5000 mile accumulation point are listed in Table 14. Each value in the table represents an average of 6 tests (2 duplications x 3 fuels).

While some of the vehicles do not show a continuous increase in aldehyde emissions with mileage, they all show an increase in individual and total aldehyde emissions from the initial to the final mileage test point. It is interesting to note that, of the vehicles tested, the Mercury Marquis had the highest aldehyde emissions; yet these aldehyde emissions were the least affected by mileage. Total aldehyde emissions for the Mercury Marquis, from the initial to the final test point, increased by roughly a factor of 2 as compared to a factor of 30 for the Ford Pinto. In general, THC emissions also increased with mileage but not to the same extent. In all cases, there was less than a two fold increase in THC emissions. The results are shown graphically in Figure 1.

These results give an indication of how aldehyde emissions vary during the first 15,000 miles. An important issue that remains is how the aldehyde emissions behave after extended mileage (i.e. 50,000 + miles). Does the trend apparent during the first 15,000 miles continue over the life of the vehicle with aldehyde emissions continually increasing with mileage? In an attempt to determine this, the data in Table 14 were extrapolated to higher mileages (50,000, 72,866 and 100,000 miles). This was done by performing a linear regression with each vehicle using the three 5000 mile accumulation points. Since certification data are collected at 4000 miles, the initial

point was not used. The linear regression parameters and extrapolated high mileage emissions can be found in Table 15. Deterioration factors were also determined with the available data and with each extrapolated high mileage point. The deterioration factors can be found in Table 16. The deterioration factors can be used with low mileage aldehyde data to project aldehyde emissions at high mileages. These projected emissions can then be compared to actual high mileage aldehyde emissions. Available high mileage aldehyde data are limited to that generated by GM. These data are presented in Table 2. The high mileage vehicles used by GM were equipped with oxidation catalysts; the average odometer reading was 72,866 miles. Using Table 4, the average total aldehyde emissions from low mileage vehicles equipped with oxidation catalysts are 3.26 mg/km. This number, multiplied by the deterioration factor calculated for 72,866 miles will give the projected aldehyde emissions at 72,866 miles. This can then be compared to the actual aldehyde emissions found by GM. The projected value is 39 mg/km, compared to the actual maximum value of 25 mg/km found by GM; however, GM used the MBTH method to generate their data. GM estimates that the MBTH aldehyde values for auto exhaust range from 10 to 50% low and average about 25% low (11). Taking this into consideration and adjusting the actual maximum value by 25% gives 31 mg/km, still lower than the projected value. If the average low mileage value generated by GM (12.4 mg/km) is multiplied by the same deterioration factor, the projected high mileage value then becomes 148 mg/km, much higher than the actual high mileage factor of 25 mg/km. Using the three-way catalyst-equipped vehicle data in Table 15, the average extrapolated aldehyde levels at 50,000 and 100,000 miles are 18.32 and 36.20 mg/km, respectively. These levels far exceed those found for low mileage three-way catalyst-equipped vehicles under malfunction conditions. It appears that aldehyde emissions from catalyst-equipped vehicles do not increase at a constant rate with increasing mileage; it is not certain with the available data whether the aldehyde emissions, 1) level off at some point, or 2) continue to increase with mileage but at a slower rate than indicated by the study. In either case, the projected high mileage aldehyde emissions from catalyst-equipped vehicles are generally less than aldehyde emissions found from non-catalyst-equipped vehicles.

The same type of analysis can be performed with THC emissions. Average THC emissions for the GM high mileage vehicles are 396 mg/km. From Table 4, the average THC emissions from low mileage vehicles equipped with oxidation catalysts are 255 mg/km. The projected THC emissions at 72,866 miles are 663 mg/km, much higher than the actual THC emissions of 396 mg/km at that mileage. These results are similar to those found for the aldehydes. It should be noted that an EPA sponsored study is presently underway to investigate aldehyde emissions from high mileage catalyst-equipped vehicles.

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Appendix

The following is a summary of the DNPH procedure used to collect and measure mobile source emissions of aldehydes and ketones. A more complete description can be found in the EPA report, "Analytical Procedures for Characterizing Unregulated Pollutant Emissions From Motor Vehicles" by Harry E. Dietzmann, et al. (5).

The collection of aldehydes (formaldehyde, acetaldehyde, isobutyraldehyde and hexanaldehyde) and ketones (acetone and methylethylketone) is accomplished by bubbling CVS diluted exhaust through glass impingers containing 2, 4-dinitrophenylhydrazine (DNPH) in dilute hydrochloric acid. The aldehydes and ketones (also known as carbonyl compounds) react with the DNPH to form their respective phenylhydrazone derivatives. These derivatives are insoluble or only slightly soluble in the DNPH/HCl solution and are removed by filtration followed by pentane extractions. The filtered precipitate and the pentane extracts are combined and then the pentane is evaporated in a vacuum oven. The remaining dried extract contains the phenylhydrazone derivatives. The extract is dissolved in a quantitative volume of toluene containing a known amount of anthracene as an internal standard. A portion of this dissolved extract is injected into a gas chromatograph and analyzed using a flame ionization detector. Acetone, acrolein and propionaldehyde are not resolved from each other under normal gas chromatographic operating conditions and all three are reported together as acetone.

FIGURE 1
EFFECTS OF MILEAGE ACCUMULATION

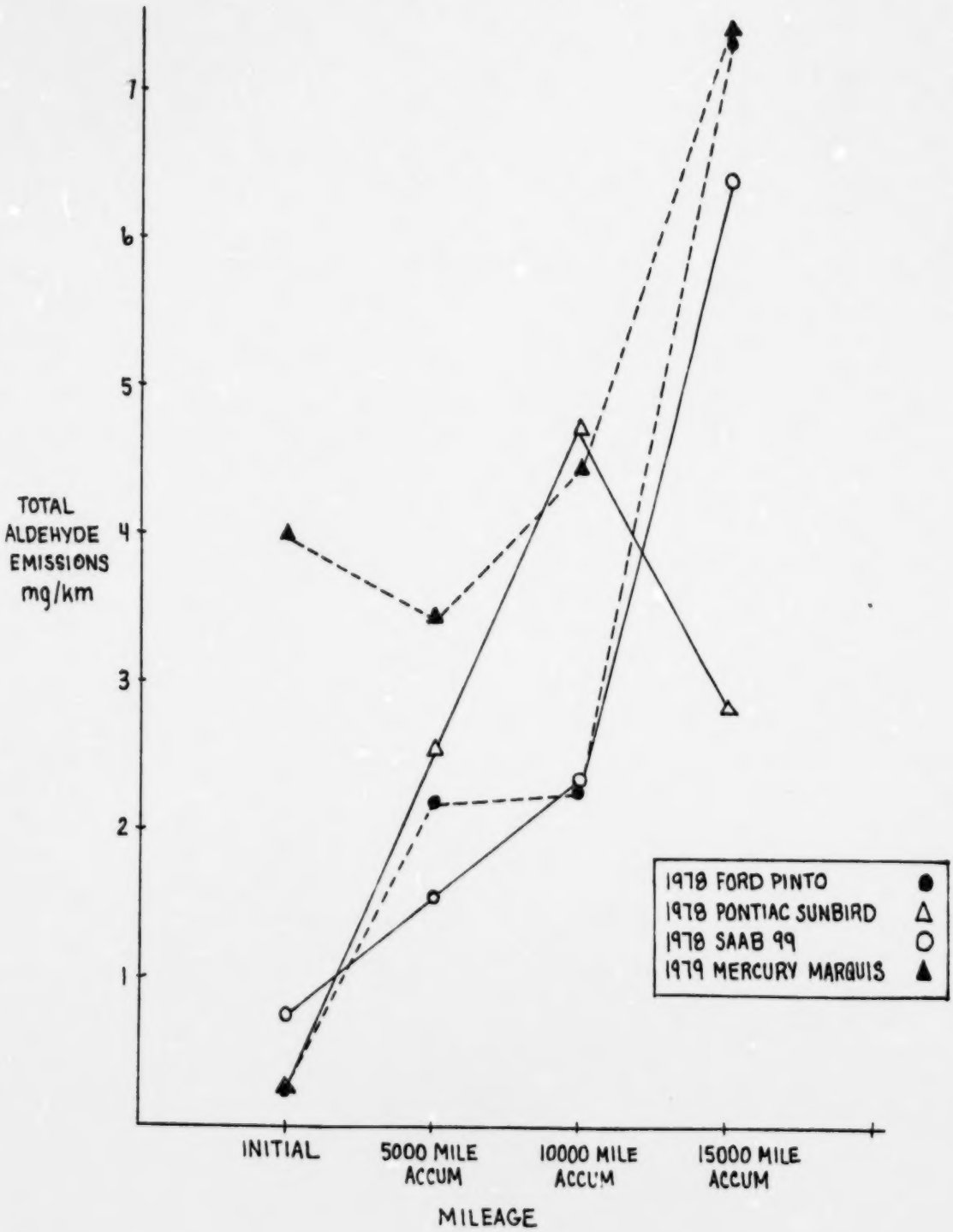


TABLE I

CHEMICAL STRUCTURES OF THE ALDEHYDES

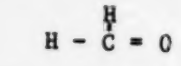
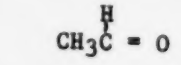
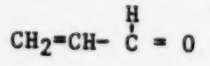
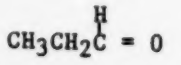
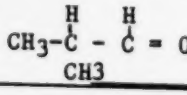
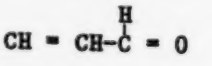
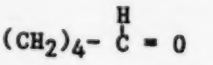
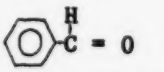
ALDEHYDE	STRUCTURE
FORMALDEHYDE	
ACETALDEHYDE	
ACROLEIN	
PROPIONALDEHYDE	
ISOBUTYRALDEHYDE	
CROTONALDEHYDE	
HEXANALDEHYDE	
BENZALDEHYDE	

TABLE 2
GM MILEAGE DATA

Vehicle* No.	Odometer	FTP HC Emissions mg/km	Aldehyde Emissions (3 tests) mg/km	Range
14	71,319	360	14.3	11.2 - 15.5
15	74,629	547	23.6	21.1 - 26.7
17	76,099	273	9.3	7.5 - 11.8
18	69,417	404	24.9	21.1 - 28.6

*1973 Oldsmobiles equipped with 1975 emission control systems

Mileage Effects

Type of Vehicle	Model Year	Range of Aldehyde Emissions mg/km
Non-Catalyst	1970-1974	49.7 - 124.3
Low-Mileage w/catalyst	1975	6.2 - 18.6
High-Mileage w/catalyst (69,417-76,099 miles)	1975*	7.5 - 28.6
Diesel	----	12.4 - 37.3

*1975 emission control systems on 1973 Oldsmobile

Exhaust Aldehyde Emissions of Special Cars

Total Aldehydes - mg/km*

Category	Oxidation Catalyst 110,000km	Dual and 3-way Catalysts	Stratified-Charge Engine	Diesel Engine	Rotary Engine**
No. of cars	4	5	5	5	2
Aldehyde min	9.3	3.1	23.0	13.7	59.04
max	24.9	4.4	74.0	37.3	315.10
avg.	18.02	3.1	55.9	21.13	187.07

*Driving cycle 1975 FTP
**Also equipped with thermal reactor

Table 3

VW Aldehyde Data

Vehicle	Catalyst	Secondary Air	Total Aldehydes mg/km
1972 Beetle	No	No	21.13
1975 Beetle	No	Yes	45.99
1975 Dasher	No	Yes	68.37
1975 Dasher	Yes	Yes	14.29
1975 Beetle	Yes	No	6.84
1975 Scirocco	Yes	Yes	6.22
1976 Beetle	Yes	No	4.97
1976 Rabbit	Yes	Yes	4.35

Average Aldehyde Emission Factor
Catalyst: 7.33 mg/km
Non-Catalyst: 45.16 mg/km

TABLE 4
FTP ALDEHYDE EMISSIONS (mg/km)
LIGHT DUTY CATALYST-EQUIPPED GASOLINE VEHICLES

	FORM-ALDEHYDE	ACET-ALDEHYDE	ACETONE[a]	ISOBUTYR-ALDEHYDE	CROTON-ALDEHYDE	HEXAN-ALDEHYDE	BENZ-ALDEHYDE	TOTAL ALDEHYDES	THC	TOTAL AS % OF THC
'77 Olds Cutlass[b]	2.60	0.40	---	3.80	---	---	---	6.80	240	2.8
'77 VW Rabbit[b]	0.40	---	---	2.60	---	---	---	3.00	140	2.1
'78 Chev. Malibu[c]	1.10	0.305	0.05	0.00	---	0.11	---	1.565	307.5	0.5
'78 Ford Granada[c]	2.01	0.14	0.05	0.00	---	0.15	---	2.35	265	0.9
'78 Ford MustangII[c]	1.58	0.63	0.15	0.03	---	0.18	---	2.57	323	0.8
'78 Ford Pinto[d] 3-way + OX Catalyst	0.79	0.00	0.035	0.00	---	0.09	---	0.915	67.5	1.4
'78 Pont. Sunbird[d] 3-way Catalyst	0.35	0.06	0.03	0.00	---	0.11	---	0.55	155.0	0.35
'78 Saab 99[d] 3-way Catalyst	0.085	0.29	0.00	0.00	---	0.00	---	0.375	100.0	0.38
'79 Merc. Marquis[e] 3-way + OX Catalyst	2.41	0.11	0.01	0.08	---	0.00	---	2.61	127.5	2.0
Emission Factor Range	0.085-2.60	0.00-0.63	0.00-0.15	0.00-3.80	---	0.00-0.18	0.375-6.80	67.5-307.5	0.35-2.8	
Average Emission Factor w/o 3-way	1.54	0.37	0.08	1.29	---	0.15	---	3.26	255	1.3
Average Emission Factor with 3-way	1.26	0.24	0.05	0.72	---	0.09	---	2.36	192	1.2

__ No Valid Data Available

[a] includes acrolein and propanal.

[b] Data from "Characterization of Sulfates, Odor, Smoke, POM and Particulates from Light and Heavy Duty Engines-Part IX," EPA-460/3-79-007, June 1979, pg. 196.

[c] Data from "Regulated and Unregulated Exhaust Emissions from Malfunctioning Non-Catalyst and Oxidation Catalyst Gasoline Automobiles", EPA-460/3-80-003, January 1980, Appendix C.

[d] Data from "Regulated and Unregulated Exhaust Emissions from Malfunctioning Three-Way Catalyst Gasoline Automobiles", EPA-460/3-80-004, January 1980, Appendix C.

[e] Data from "Regulated and Unregulated Exhaust Emissions from a Malfunctioning Three-Way Catalyst Gasoline Automobile", EPA-460/3-80-005, January 1980.

TABLE 5

FTP ALDEHYDE EMISSIONS (mg/km)
LIGHT-DUTY NON-CATALYST GASOLINE VEHICLES

VEHICLE	FORM-ALDEHYDE	ACET-ALDEHYDE	ACETONE[a]	ISOBUTYR-ALDEHYDE	CROTON-ALDEHYDE	HEXAN-ALDEHYDE	BENZ-ALDEHYDE	TOTAL ALDEHYDES	THC	TOTAL AS % OF THC
1972[b] Olds 98	---	---	---	---	---	---	---	111.9	1069	10.5
1970[b] Volkswagon	---	---	---	---	---	---	---	59.7	1187	5.0
1971[b] Ford Galaxie	---	---	---	---	---	---	---	105.7	3456	3.1
1971[b] Plymouth FuryIII	---	---	---	---	---	---	---	111.9	2200	5.1.
1972[b] Ford Torino	---	---	---	---	---	---	---	62.1	1423	4.4
1970[b] Chev.Impala	---	---	---	---	---		---	93.2	2449	3.8
1970[b] Pontiac	---	---	---	---	---	---	---	205.1	3679	5.6
1971[b] Chev. Vega	---	---	---	---	---	---	---	74.6	3381	2.2
1973[b] Ford Torino	---	---	---	---	---	---	---	111.9	1666	6.7
1973[b] Chev. Impala	---	---	---	---	---	---	---	149.2	1318	11.3
1970 Olds[c] Delta 88	31.87	4.46	0.71	---	---	0.00	---	37.04	1430	2.6
1970[c] Dodge Challenger	9.31	3.45	0.00	---	---	0.00	---	12.76	1710	0.7
1970 Chevy[c] Monte Carlo	66.77	6.74	0.00	---	---	0.00	---	73.51	2050	3.6
1970 Ford[c] Fairlane	21.29	1.42	0.23	---	---	0.00	---	22.94	2350	1.0
Emission Factor Range	9.31-66.77	142-6.74	0.00-0.71	---	---	---	---	12.76-205.1	1069-3679	0.7-10.5
Average Emission Factor	32.31	4.02	0.24	---	---	0.00	---	87.97	2098	4.2

--- No Data Available

[a] Includes acrolein and propanal

[b] Data from "Aldehyde and Reactive Organic Emissions from Motor Vehicles, Part II", APTD-1568b, March 1973.

[c] Data from "Unregulated Exhaust Emissions From Non-Catalyst Baseline Cars Under Malfunction Conditions", Contract No. 68-03-2884, 1981.

TABLE 6
FTP ALDEHYDE EMISSIONS (mg/km)
LIGHT-DUTY DIESEL VEHICLES

	FORM-ALDEHYDE	ACET-ALDEHYDE	ACETONE[a]	ISOBUTYR-ALDEHYDE	CROTON-ALDEHYDE	HEXAN-ALDEHYDE	BENZ-ALDEHYDE	TOTAL ALDEHYDES	THC	TOTAL AS % OF THC
'74 Perkins[b] 6-247	38.17	10.50	5.31	13.00	---	0.00	---	66.98	450.	14.9
'74 Peugot[b] 204D	11.25	4.28	3.01	8.75	---	0.00	0.00	27.29	690.	4.0
'75 Mercedes[b] 220D Comprex	2.52	1.00	8.37	11.08	---	0.47	0.00	23.44	110.	21.3
'75 Mercedes[b] 240D	3.96	1.13	1.47	2.19	---	0.00	0.00	8.75	180.	4.9
'75 Mercedes[b] 300D	3.80	1.11	9.41	0.00	---	0.00	---	14.32	100.	14.3
'76 Cutlass[c] Diesel	15.80	6.50	35.70	18.50	---	---	---	76.50	470.	16.3
'77 Rabbit[c] Diesel	16.00	5.00	2.60	16.00	---	---	---	39.60	230.	17.2
Emission Factor Range	2.52-38.17	1.00-10.50	1.47-35.70	0.00-18.50	---	0.00-0.47	---	8.75-76.50	100-690	4.0-21.3
Average Emission Factor	13.07	4.22	9.41	9.93	---	0.07	---	36.70	381.6	11.5

--- No Valid Data Available
[a]Includes Acrolein and Propanal
[b]Data from "Investigation of Diesel-Powered Vehicle Emissions VII", EPA-460 /3-76-034, Feb. 1977, Pg. 172.
[c]Data from "Characterization of Sulfates, Odor, Smoke, POM, and Particulates from Light and Heavy Duty Engines-Part IX", EPA-460/3-79-007, June 1979.

TABLE 7
ALDEHYDE EMISSIONS
HEAVY-DUTY VEHICLES[a,b]

		FORM-ALDEHYDE	ACET-ALDEHYDE	ACETONE[c]	ISOBUTYR-ALDEHYDE	CROTON-ALDEHYDE	HEXAN-ALDEHYDE	BENZ-ALDEHYDE	TOTAL ALDEHYDES	THC	TOTAL AS % OF THC
Chevrolet Gasoline	mg/kw-hr	104.75	18.09	18.40	13.79	---	42.83	---	197.86	3340[d]	5.9
	mg/kg fuel	228.88	39.53	40.20	30.13	---	93.58	---	432.32		
	mg/km[e]	92.82-59.11	16.03-10.21	16.30-10.38	12.22-7.78	---	37.95-24.17	---	175.32-111.65		
	Avg. mg/km	75.97	13.12	13.34	10.00	---	31.06	---	143.49		
Mack ETAY(B)673A Diesel	mg/kw-hr	16.59	0.94	---	31.79	---	20.18	---	69.50	638	10.9
	mg/kg fuel	166.84	3.78	---	128.12	---	83.87	---	282.61		
	mg/km[e]	31.17-19.85	1.76-1.12	---	59.75-38.05	---	39.11-24.91	---	131.79-83.93		
	Avg. mg/km	25.51	1.44	---	48.90	---	32.01	---	107.86		
Caterpillar 3208/EGR Diesel	mg/kw-hr	49.90	21.85	12.55	3.18	---	13.98	---	101.46	1559	6.5
	mg/kg fuel	223.76	97.98	56.28	14.24	---	62.70	---	454.96		
	mg/km[e]	104.35-66.46	45.69-29.10	26.25-16.72	6.64-4.23	---	29.24-18.62	---	212.17-135.13		
	Avg. mg/km	85.41	37.40	21.49	5.43	---	23.93	---	173.66		
	DIESEL AVG. mg/kw-hr	33.25	11.40	6.27	17.48	---	17.08	---	85.48	1099	7.8
	DIESEL AVG. mg/km	55.46	19.42	10.75	27.17	---	27.97	---	140.77		

New York City-Los Angeles Usage

---No Valid Data Available

[a]Based on Weighting Factors Derived form 13-Mode FTP.

[b]Data from Characterization of Sulfates, Odor, Smoke, POM and Particulates From Light-and Heavy-Duty Engines Part IX, EPA-460/3-79-007, June 1979.

[c]Includes Acrolein and Propanal

[d]Based on 23-Mode FTP

[e]Emission Factor Data Derived using T.M. Baines, et al., "Heavy-Duty Diesel Particulate Emission Factors", Journal of the Air Pollution Control Association, Volume 29, No. 6; June 1979.

TABLE 8

SUMMARY OF VEHICULAR ALDEHYDE AND HYDROCARBON EMISSIONS

	HC mg/km	Aldehydes mg/km	Form-Aldehyde mg/km	Ald/HC%	No. of Cars Engines Tested
LD Gasoline Non-Catalyst	2098	88	32[b]	4	14
LD Gasoline-Catalsyt including 3-way catalyst vehicles	192	2	1	2	9
excluding 3-way catalyst vehicles	255	3	2	2	5
LD Diesel	319	37	13	12	7
HD Diesel[a]	1099	86	33	8	2
	---	141	55	---	2
HD Gasoline[a]	3340	198	105	6	1
	---	143	76	---	1

[a]mg/kw-hr
[b]based on four vehicles

Table 9
MALFUNCTION CONFIGURATION
ALDEHYDE EMISSIONS

		AVERAGE EMISSION RATE, mg/km							
		UNMODIFIED FTP	12 PERCENT MISFIRE	DISABLED EGR	RICH BEST IDLE	HIGH OIL CONSUMPTION	DISABLED O_2 SENSOR	DISABLED EGR AND O_2	DISABLED O_2 SENSOR AND AIR PUMP
	Formaldehyde	9.71	75.24	11.055	12.62	21.725	---	---	---
	Acetaldehyde	2.345	31.04	2.205	2.23	7.23	---	---	
	Acetone[a]	0.765	10.00	1.01	0.075	1.27	---	---	---
'77 AMC PACER	Isobutyraldehyde.	0.00	0.00	0.00	0.00	0.00	---	---	---
[b]	Crotonaldehyde	---	---	---	---	---	---	---	
W/O Catalyst	Hexanaldehyde	0.19	1.915	0.165	0.08	0.435	---	---	---
	Benzaldehyde	---	---	---	---	---	---	---	---
	Total Aldehydes	13.01	118.20	14.44	15.01	30.66	---	---	---
	THC	725.0	10820.	785.0	805.0	1115.	---	---	---
	Total as % of THC	1.8	1.1	1.8	1.9	2.7	---	---	---
	Formaldehyde	1.10	2.29	2.22	1.305	0.165	---	---	
	Acetaldehyde	0.305	2.115	0.88	0.405	0.20	---	---	
	Acetone[a]	0.05	0.295	0.265	0.00	0.09	---	---	---
'78 Chev. Malibu	Isobutyraldehyde	0.00	0.00	0.00	0.03	0.03	---	---	---
[b]	Crotenaldehyde	---	---	---	---	---	---	---	---
	Hexanaldehyde	0.11	0.10	0.105	0.04	0.095	---	---	---
	Benzaldehyde	---	---	---	---	---	---	---	---
	Total Aldehydes	1.57	4.8	3.47	1.78	0.58	---	---	---
	THC	307.5	1780	335	1080	570	---	---	---
	Total as % of THC	0.51	0.27	1.04	0.16	0.10	---	---	---
	Formaldehyde	0.79	0.00	---	---	---	0.00	0.475	0.315
	Acetaldehyde	0.00	0.0	---	---	---	0.015	0.00	0.615
	Acetone[a]	0.035	0.00	---	---	---	0.135	0.00	0.00
'78 Ford Pinto	Isobutyraldhyde	0.00	0.00	---	---	---	0.00	0.00	0.09
[c]	Crotonaldehyde	---	---	---	---	---	---	---	---
3-Way+OX	Hexanaldehyde	0.09	0.00	---	---	---	0.255	0.00	0.00
Catalyst	Benzaldehyde	---	---	---	---	---	---	---	---
	Total Aldehydes	0.92	0.00	---	---	---	0.41	0.48	1.02
	THC	67.5	380	---	---	---	95	115	1160
	Total as % of THC	1.4	0.00	---	---	---	0.43	0.42	0.09

---No Valid Data Available ---not measured

[a]Includes Acrolein and Propanal

[b]Data from "Regulated and Unregulated Exhaust Emissions From Malfunctioning Non-Catalyst and Oxidation Catalyst Gasoline Automobiles", EPA-460/3-80-003, January 1980, Appendix C.

[c]Data from "Regulated and Unregulated Exhaust Emissions From Malfunctiong Three-way Catalsyt Gasoline Automobiles", EPA-460/3-80-004, January 1980, Appendix C.

TABLE 10
MALFUNCTION CONFIGURATION ALDEHYDE EMISSIONS
(CONT.)

		AVERAGE EMISSIONS RATE, mg/km							
		UNMODIFIED FTP	12 PERCENT MISFIRE	DISABLED O_2 SENSOR	DISABLED EGR AND O_2	HIGH OIL CONSUMPTION	DISABLED O_2 SENSOR AND RICH BEST IDLE	12 PERCENT MISFIRE AND AIR TO BYPASS	DISCONNECTED CTS
'78 Pontiac Sunbird[b] 3-WAY Catalyst	Formaldehyde	0.35	0.39	0.74	1.675	0.18	---	---	---
	Acetaldehyde	0.06	0.76	2.005	4.695	0.00	---	---	---
	Acetone[a]	0.03	0.045	0.40	0.00	0.025	---	---	---
	Isobutyraldehyde	0.00	0.00	0.00	0.00	0.00	---	---	---
	Crotonaldehyde	---	---	---	---	---	---	---	---
	Hexanaldehyde	0.11	0.00	0.355	0.00	0.00	---	---	---
	Benzaldehyde	---	---	---	---	---	---	---	---
	Total Aldehydes	0.55	1.20	3.50	6.37	0.21	---	---	---
	THC	155	1050	1470	1400	240	---	---	---
	Total As % of THC	0.35	0.11	0.24	0.46	0.09	---	---	---
'78 Saab 99[b] 3-Way Catalyst	Formaldehyde	0.085	0.00	0.00	---	0.01	0.27	---	---
	Acetaldehyde	0.29	0.00	0.00	---	0.29	2.47	---	---
	Acetone[a]	0.00	0.00	0.23	---	0.00	0.075	---	---
	Isobutyraldehyde	0.00	0.00	0.17	---	0.02	0.00	---	---
	Crotonaldehyde	---	---	---	---	---	---	---	---
	Hexanaldehyde	0.00	0.00	0.00	---	0.00	0.00	---	---
	Benzaldehyde	---	---	---	---	---	---	---	---
	Total Aldehydes	0.38	0.00	0.40	---	0.32	2.82	---	---
	THC	100	2150	710		130	815	---	---
	Total As % of THC	0.38	0.00	0.06	---	0.25	0.35	---	---
'79 Mercury Marquis[c] 3-WAY Catalyst	Formaldehyde	2.41	---	0.00	1.265	---	---	0.91	0.22
	Acetaldehyde	0.11	---	0.12	0.41	---	---	0.655	1.17
	Acetone[a]	0.01	---	0.00	0.145	---	---	0.00	1.095
	Isobutyraldehyde	0.08	---	0.13	1.095	---	---	0.275	0.00
	Crotonaldehyde	---	---	---	---	---	---	---	---
	Hexanaldehyde	0.00	---	0.00	0.00	---	---	0.00	0.00
	Benzaldehyde	---	---	---	---	---	---	---	---
	Total Aldehydes	2.61	---	0.25	2.92	---	---	1.85	2.49
	THC	127.5	---	105	1485	---	---	1725	1795
	Total As % of THC	2.05	---	0.24	0.20	---	---	0.11	0.14

---No Valid Data Available ___Not Measured

[a]Includes Acrolein and Propanal

[b]Data From "Regulated and Unregulated Exhaust Emissions From Malfunctioning Three-Way Catalyst Gasoline Automobiles", EPA-460/3-80-004, January 1980, Appendix C.

[c]Data from "Regulated and Unregulated Exhaust Emissions From a Malfunctioning Three-Way Catalyst Gasoline Automobile", EPA-460/3-80-005, January 1980, Appendix C.

TABLE 11
FUEL VARIATION ALDEHYDE EMISSIONS

	FUEL	FORM-ALDEHYDE	ACET-ALDEHYDE	ACETONE[a]	ISOBUTYL-ALDEHYDE	CROTON-ALDEHYDE	HEXAN-ALDEHYDE	BENZ-ALDEHYDE	TOTAL ALDEHYDES	THC	TOTAL AS % OF THC
MERCEDES 240D[b,d]											
	EM-238-F	4.70	1.00	3.60	0.56	---	0.75	---	10.61	120	8.8
	EM-239-F	5.00	0.69	3.10	0.60	---	1.20	0.00	10.59	190	5.6
	EM-240-F	2.50	0.087	0.31	2.90	---	1.30	---	7.10	90	7.9
	EM-241-F	7.00	1.30	2.60	1.20	---	2.10	---	14.20	200	7.1
	EM-242-F	5.4	0.57	1.20	4.20	---	2.20	---	13.57	120	11.3
VW RABBIT DIESEL[b,d]											
	EM-238-F	6.00	1.20	5.50	0.73	---	3.90	---	17.33	180	9.6
	EM-239-F	5.10	0.39	1.10	1.30	---	0.84	---	8.73	200	4.4
	EM-240-F	4.00	0.41	0.46	1.20	---	2.90	---	8.97	170	5.3
	EM-241-F	19.00	4.81	8.40	2.30	---	3.90	---	38.40	710	5.4
	EM-242-F	6.20	0.86	1.60	1.80	---	1.00	0.00	11.46	200	5.7
DETROIT DIESEL ALLISON 6V-71[c,e]											
	EM-238-F	137.53	81.40	182.44	322.78	---	11.23	---	735.38	500	147.1
	EM-239-F	72.49	0.70	5.16	0.00	---	22.31	---	100.66	710	14.2
	EM-240-F	47.68	24.56	28.89	96.79	---	7.22	0.00	205.14	990	20.7
	EM-241-F	94.78	23.69	85.02	76.66	---	41.81	0.00	321.96	850	37.9
	EM-242-F	63.35	7.44	12.67	28.92	---	13.08	0.00	125.46	690	18.2
CATERPILLAR 3208[c,e]											
	EM-238-F	100.55	38.21	24.49	52.28	---	16.09	---	236.62	1240	19.1
	EM-239-F	92.89	14.60	35.83	35.17	---	23.89	---	202.38	1110	18.2
	EM-240-F	126.54	39.37	70.30	140.60	---	11.25	---	388.06	1620	24.0
	EM-241-F	166.18	42.54	35.89	53.18	---	9.31	---	307.10	1880	16.3
	EM-242-F	67.39	27.23	25.87	25.87	---	4.83	---	151.19	1100	13.7

[a]Includes Acetone, Acrolein and Propanal
[b]Emission Factors, mg/km
[c]Aldehyde Emission Factors, mg/hp hr at 101.3kPA and 21°C, THC Emission Factors, mg/hp hr
[d]Data from "Characterization of Gaseous and Particulate Emissions from Light-Duty Diesels Operated on Various Fuels", EPA 460/3-79-008, July 1979.
[e]Data from "Characterization of Diesel Gaseous and Particulate Emissions", Contract No. 68-02-1777, Sept. 1977 (unpublished)

TABLE 12
TEMPERATURE VARIATION ALDEHYDE EMISSIONS
(mg/km)

TEMPERATURE F (C)	0 (-18)	20 (-7)	40 (4)	60 (16)	70 (21)	80 (27)	80AC (27)	90 (32)	90AC (32)	110 (43)	110AC (43)
1972 Chev. Impala	92.64	103.31	79.55	100.81	109.84	116.60	127.54	124.29	118.12	136.93	98.40
W/O Catalyst	100.41	99.84	85.03	90.07	88.19	82.36	84.41	77.15	77.51	88.30	89.39
Ave	96.53	101.57	82.29	95.44	99.01	99.48	105.97	100.72	97.82	112.64	93.89
1974 Chev. Impala	244.21	205.56	202.87	139.19	142.30	148.10	•	158.05	•	198.97	•
W/O Catalyst	184.07	175.04	285.26	260.27	260.16	211.13	•	176.41	•	200.34	•
	•	•	•	263.36	•	•	•	•	•	•	•
Ave	214.14	190.80	244.07	220.94	201.23	179.61	•	167.23	•	199.66	•
1977 Honda Civic	277.22	322.44	192.83	276.38	141.30	170.94	•	152.50	•	170.83	•
49 State	270.85	247.61	272.30	214.57	235.88	179.38	•	175.55	•	195.23	•
W/O Catalyst Ave	274.04	285.03	232.56	245.48	188.59	175.15	•	164.03	•	183.03	•
1977 Ford LTD	149.97	106.10	99.96	110.32	118.96	91.97	95.03	70.01	82.37	81.10	81.76
49 State	113.36	84.46	84.50	96.33	111.69	107.41	109.25	66.77	75.16	66.94	81.24
	70.11	•	•	•	•	•	•	•	•	•	•
Ave	111.15	95.63	92.23	103.32	115.32	99.60	102.14	68.39	78.76	74.02	81.50
1977 Plymouth Fury	121.31	114.37	102.92	81.47	82.76	82.89	•	88.85	•	62.75	•
49 State	108.08	•	130.41	102.24	92.88	103.21	•	100.42	•	85.99	•
	•	•	•	65.25	46.86	•	•	•	•	•	•
Ave	114.69	114.37	116.66	82.99	74.17	93.05	•	94.64	•	74.37	•
1978 Buick V6	77.75	118.76	62.98	146.89	171.76	137.63	138.58	136.51	135.71	157.31	127.56
Turbocharged	119.52	111.38	64.40	60.03	66.36	70.22	95.60	115.13	90.28	93.34	123.17
	111.41	•	•	•	•	•	•	•	•	•	•
Ave	102.89	115.07	63.69	103.36	119.06	103.92	117.09	125.82	112.99	125.32	125.37
1977 Plymouth Fury	74.91	57.53	56.85	66.81	69.10	45.34	47.37	48.10	44.16	35.37	46.53
Calif.	77.82	57.15	45.19	82.87	61.15	40.72	45.98	47.92	55.84	50.31	58.01
	•	•	•	•	•	•	•	•	•	•	49.08
Ave	76.37	57.34	51.02	74.74	65.12	43.03	44.68	47.92	50.01	42.84	51.21
1978 Chevy. Sc.	69.83	63.03	64.65	49.33	46.24	44.20	39.12	35.21	37.25	30.95	54.99
Calif.	78.53	93.15	61.59	50.74	57.35	56.68	58.82	59.14	63.56	40.81	50.88
Ave	73.95	79.09	63.12	50.04	51.79	50.44	48.97	47.18	50.40	35.88	52.94
1978 Ford Pinto	73.14	57.17	68.17	63.59	50.61	33.22	31.92	50.37	38.43	42.34	45.67
Calif.-3 Way	69.83	61.14	45.40	67.95	77.29	47.07	57.02	44.46	31.64	34.56	31.08
	85.00	•	•	•	•	•	•	29.95	•	•	81.20
Ave	75.99	59.16	56.79	65.77	63.95	40.15	44.47	41.59	35.04	38.45	52.65
1978 VW Rabbit	51.14	48.11	57.80	97.28	98.90	70.15	•	78.18	•	83.62	•
Calif. FUINJ	60.66	76.32	57.50	169.48	58.71	66.34	•	53.00	•	53.16	•
Ave	55.90	62.22	57.65	133.38	78.81	68.25	•	65.59	•	68.39	•
1979 Dodge Aspen	116.80	111.75	96.35	77.24	73.22	80.79	129.1-	141.14	99.15	115.22	105.58
Calif.	79.73	71.46	72.15	96.45	86.40	90.93	89.07	82.75	101.20	113.03	138.13
Ave	98.26	91.61	84.25	86.85	84.81	85.86	109.10	111.95	100.17	114.12	121.85
1980 Mercury	•	48.12	62.25	64.75	59.06	41.49	54.96	57.73	41.08	62.87	44.93
Prototype	•	52.49	•	•	75.67	37.84	•	•	•	39.42	57.72
Ave	•	50.31	62.25	64.75	67.37	39.66	54.96	57.73	41.08	51.14	51.33
1980 Buick Regal	•	59.19	52.86	23.01	41.69	50.17	19.19	42.79	51.15	56.78	54.36
Prototype	•	40.76	•	•	26.83	53.78	•	•	•	39.16	102.01
Ave	•	49.97	52.86	23.01	34.26	51.96	19.19	42.79	51.15	47.97	78.18
Datson Prototype	53.02	100.90	76.32	57.06	45.56	40.17	•	150.27	•	64.22	•
	62.66	49.97	57.01	95.64	75.38	112.41	•	125.83	•	28.04	•
	•	•	80.84	•	•	•	•	•	•	•	•
Ave	57.84	75.38	71.39	76.35	60.47	76.29	•	138.05	•	46.13	•

TABLE 13

FTP ALDEHYDE EMISSIONS FROM 1979 OLDSMOBILE DIESEL DELTA 88 WITH AND WITHOUT A CATALYST (mg/km)

TEST NO.	CATALYST	FORM-ALDEHYDE	ACET-ALDEHYDE	ACETONE[a]	ISOBUTYR-ALDEHYDE	CROTON-ALDEHYDE	HEXAN-ALDEHYDE	BENZ-ALDEHYDE	TOTAL ALDEHYDES	THC	TOTAL AS % OF THC
1	NONE	3.4	1.2	0.1	2.1	0.0	0.0	0.0	6.8	260	2.6
3	NONE	6.1	0.9	0.8	0.0	---	0.0	---	7.8	170	4.6
5	PTX-516	1.1	0.6	0.0	0.0	0.0	0.0	---	1.7	90	1.9
7	PTX-516	2.4	0.7	0.0	0.0	0.0	0.0	---	3.1	90	3.4
9-1	None	4.2	0.9	0.0	0.0	0.0	0.0	---	5.1	270	1.9
10-1	UOP-103	3.1	0.7	0.0	0.0	0.0	0.0	0.0	4.4	130	3.4
11-1	UOP-103	2.9	0.9	0.4	0.0	0.0	0.0	0.0	4.2	40	10.5
12-1	NONE	4.9	0.1	0.0	0.0	0.0	0.0	0.0	5.0	280	1.8
13-1	UOP-99	4.5	0.0	0.0	0.0	---	0.0	---	4.5	210	2.1
14-1	UOP-99	5.3	0.0	0.0	1.6	0.0	0.0	---	6.9	180	3.8
					(AVERAGE EMISSIONS, mg/km)						
1,3,9-1,12-1	NONE	4.7	0.8	0.2	0.5	0.0	0.0	---	6.2	245	2.5
5,7	PTX-516	1.8	0.7	0.0	0.0	0.0	0.0	---	2.5	90	2.8
10-1,11-1	UOP-103	3.0	0.8	0.2	0.3	0.0	0.0	0.0	4.3	85	5.1
13-1,14-1	UOP-99	4.9	0.0	0.0	0.8	---	0.0	---	5.7	195	2.9

[a]Includes Acrolein and Propanal

TABLE 14
MILEAGE COMPARISONS FOR ALDEHYDES

		Average Emission Rates, mg/km			
		Initial	After First 5000 Mile Accum	After Second 5000 Mile Accum	After Third 5000 Mile Accum
'78 Ford Pinto 3-way+OX.	Formaldehyde	0.14	1.28	1.20	1.33
	Acetaldehyde	0.09	0.14	0.15	0.36
	Isobutyraldehyde	0.01	0.78	0.91	5.18
	Hexanaldehyde	ND	ND	ND	0.48
	Total Aldehydes	0.24	2.20	2.26	7.35
	THC	160	200	260	230
	Total as % of THC	0.2	1.1	0.9	3.2
'78 Pontiac Sunbird 3-Way	Formaldehyde	0.22	0.88	0.55	1.05
	Acetaldehyde	ND	0.25	0.58	0.33
	Isobutyraldehyde	0.05	1.44	3.46	1.17
	Hexanaldehyde	ND	ND	0.15	0.31
	Total Aldehydes	0.27	2.57	4.74	2.86
	THC	270	270	310	380
	Total as % of THC	0.1	1.0	1.5	0.8
'78 Saab 99 3-Way	Formaldehyde	0.10	0.36	0.04	0.24
	Acetaldehyde	0.55	0.14	0.27	0.64
	Isobutyradlehyde	0.05	1.16	1.99	5.00
	Hexanaldehyde	0.04	0.01	0.08	0.54
	Total Aldehydes	0.74	1.67	2.38	6.42
	THC	130	110	150	140
	Total as % of THC	0.6	1.5	1.6	4.6
'79 Merc Marquis 3-Way + OX	Formaldehyde	3.65	3.23	2.82	4.05
	Acetaldehyde	0.30	0.08	0.33	0.88
	Isobutyraldehyde	0.04	0.15	1.32	2.20
	Hexanaldehyde	ND	ND	ND	0.28
	Total Aldehydes	3.99	3.46	4.47	7.41
	THC	140	210	200	230
	Total as % of THC	2.9	1.6	2.2	3.2

Table 15

Linear Regression Parameters and Extrapolated High Mileage Emissions

Vehicles	Linear Regression Parameters			Extrapolated Emissions, mg/mk		
	Slope	Y-Intercept	Correlation Coefficient	50,000 Miles	72,866 Miles	100,000 Miles
1978 Ford Pinto						
Aldehydes	0.0005	-1.213	0.871	23.79	35.22	48.79
THC	0.003	200.	0.500	350.	418.60	500.
1978 Pontiac Sunbird						
Aldehydes	0.00003	3.10	0.123	4.60	5.29	6.10
THC	0.011	210.	0.988	760.	1011.53	1310.
1978 Saab 99						
Aldehydes	0.0005	-1.26	0.927	23.74	35.17	48.74
THC	0.003	103.33	0.721	253.33	321.93	403.33
1979 Mercury Marquis						
Aldehydes	0.0004	1.16	0.962	21.16	30.31	41.16
THC	0.002	193.33	0.655	293.33	339.06	393.33

Table 16

Deterioration Factors*

Vehicles	Deterioration Factors Using Available Data			Deterioration Factors Using Extrapolated Data		
	10,000 Miles	15,000 Miles	Average	50,000 Miles	72,866 Miles	100,000 Miles
1978 Ford Pinto						
Aldehydes	1.03	3.34	2.19	10.81	16.01	22.18
THC	1.30	1.15	1.23	1.75	2.09	2.50
1978 Pontiac Sunbird						
Aldehydes	1.84	1.11	1.47	1.79	2.06	2.37
THC	1.15	1.41	1.28	2.81	3.75	4.85
1978 Saab 99						
Aldehydes	1.43	3.84	2.63	14.22	21.06	29.19
THC	1.36	1.27	1.32	2.30	2.93	3.67
1979 Mercury Marquis						
Aldehydes	1.29	2.14	1.72	6.12	8.76	11.90
THC	0.95	1.10	1.02	1.40	1.61	1.87
Average						
Aldehdyes	1.40	2.60	2.00	8.24	11.97	16.41
THC	1.19	1.23	1.21	2.07	2.60	3.22

*Deterioration Factor = $\frac{\text{emission factor at specified mileage, mg/km}}{\text{emission factor at 5000 miles, mg/km}}$